

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**

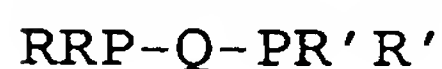
WE CLAIM:

1. A catalyst composition comprising:
  - a) A cobalt compound; and
  - b) An iron component, optionally ligated with a ligand selected from the group consisting of N-heterocycle, phosphine, and porphorine moieties.
2. The composition of Claim 1 wherein the cobalt compound is selected from cobalt salts that are reduced to the zero valence state by heat treatment in the presence of synthesis gas.
3. The composition of Claim 1 wherein the cobalt salt is selected from cobalt carboxylates and cobalt salts of mineral acids, and mixtures thereof.
4. The composition of Claim 1 wherein the cobalt compound is selected from a cobalt carbonyl.
5. The composition of Claim 4 wherein the cobalt compound is dicobalt octacarbonyl.
6. The composition of Claim 1 wherein the iron compound is an iron salt that is readily reduced to the zero valence state by heat treatment in an atmosphere of hydrogen and carbon monoxide.
7. The composition of Claim 1 wherein the iron salt is selected from the group consisting of iron carboxylates, iron salts of mineral acids, and mixtures thereof.
8. The composition of Claim 1 wherein the iron compound is an iron carbonyl or organometallic iron compound.
9. The composition of Claim 1 wherein the ligand is present and is selected from the group consisting of monodentate, bidentate, and multidentate N-heterocyclic ligands.
10. The composition of Claim 9 wherein the ligand is a diazine or a benzodiazine.

11. The composition of Claim 10 wherein the ligand is selected from the group consisting of pyrimidine, pyrazine, pyridazine, quinazoline, and quinoxaline.
12. The composition of Claim 9 wherein the ligand is a bipyridine.
13. The composition of Claim 12 wherein the ligand is selected from the group consisting of 2,2'-dipyridyl, 2,2'-bipyrimidine, 1,10-phenanthroline, di-2-pyridyl ketone, 4,4'-dimethyl-2, 2'-dipyridyl, 5,6-dimethylphenanthroline, 4,7-dimethylphenanthroline, 2,2'-biquinoline, neocuproine, and 2,2'-dipyridylamine.
14. The composition of Claim 9 wherein the ligand is a multipyridine.
15. The composition of Claim 14 wherein the ligand is selected from the group consisting of 2,4,6-tripyridyl-s-triazine, 3,6-di-2-pyridyl-1, 2,4,5-tetrazine, 2,2': 6', 2"-terpyridine, 2,3-bis (pyridyl)pyrazine, and 3-(2-pyridyl)-5,6-diphenyl-1,2,4-triazine.
16. The composition of Claim 9 wherein the ligand is a pyridine.
17. The composition of Claim 16 wherein the ligand is selected from the group consisting of pyridine, 3-hydroxypyridine, and quinoline.
18. The composition of Claim 17 wherein the ligand is a lower cost homologues of 3-hydroxypyridine and quinoline derived from coal-tar extracts.
19. The composition of Claim 9 wherein the ligand is a 2,6-pyridyl-diimine.
20. The composition of Claim 19 wherein the ligand is selected from the group consisting of 2,6-bis (N-phenyl, methylimino) pyridine and 2,6-bis[N-(2,6-diisopropylphenyl)methylimino]pyridine.
21. The composition of Claim 9 wherein the ligand is a N-heterocycle selected from the group consisting of 2,2'-

dipyridyl(DIPY), 2,2'-bipyrimidine(BPYM), and 2,4,6-tripyridyl-s-triazine(TPTZ).

22. The composition of Claim 1 wherein the ligand is present and is a tertiary diphosphine ligand of the general formula:



wherein each group R and R' independently or jointly is a hydrocarbon moiety of up to 30 carbon atoms, and Q is an organic bridging group of 2 to 4 atoms in length.

23. The composition of claim 22 wherein group R or R' each independently is an alkyl, cycloalkyl, bicycloalkyl or aryl group.

24. The composition of Claim 23, wherein group R or R' each independently has up to 20 carbon atoms.

25. The composition of Claim 22 wherein group Q is composed of carbon atoms.

26. The composition of Claim 25 wherein Q is an alkylene group of 2, 3 or 4 carbon atoms in length.

27. The composition of Claim 22, wherein both groups R and/or R' form, with the phosphorus atom, a phosphacycloalkane of from 5 to 8 atoms.

28. The composition of Claim 22, wherein the tertiary diphosphine ligand is selected from the group consisting of 1,2-bis(dimethylphosphino)ethane; 1,2-

bis(diethylphosphino)ethane; 1,2-

bis(diisobutylphosphino)ethane; 1,2-

bis(dicyclohexylphosphino)ethane; 1,2-bis(2,4,4-trimethylpentylphosphino)ethane; 1,2-

bis(diethylphosphino)propane; 1,3-

bis(diethylphosphino)propane; 1-diethylphosphino)-3-dibutylphosphino)propane; 1,2-

bis(diphenylphosphino)ethane; 1,2-

bis(dicyclohexylphosphino)ethane; 1,2-bis(2-pyridyl, phenylphosphanyl)benzene; 1,2-

bis(dicyclopentylphosphino)ethane; 1,3-bis(2,4,4-trimethylpentylphosphino)propane; 1,2-bis(diphenylphosphino)benzene; 1,2-bis(phospholano)ethane, 1,2-bis(2,5-dimethylphospholano)benzene, optically pure (R,R), (R,S), (S,S) 1,2-bis(2,5-dimethylphospholano)ethane or its racemic mixture, 1,2-bis(9-phosphabicyclononyl)ethane, 1,2-bis(2,4,4-trimethylpentylphosphino)ethane, 1,2-bis(diisobutylphosphino)ethane, 1,2-P,P'-bis(9-phosphabicyclo[3.3.1] and/or [4.2.1]nonyl)ethane, its 1,2-P,P'-propane analogue, and its 1,3-P,P'-propane analogue.

29. The composition of Claim 1 wherein the ligand is present and is a porphorine incorporating four pyrrole-type moieties in a cyclic structure.

30. The composition of Claim 29 wherein the porphorine is selected from the group consisting of octaethylporphorines and tetraphenylporphorines.

31. The composition of Claim 30 wherein the porphorine is 2,3,7,8,12,13,17,18-octaethyl-21H,23H-porphorine.

32. The composition of Claim 29 wherein the porphorine is a phthalocyanine.

33. The composition of Claim 1 wherein the iron is ligated and the molar ratio of ligand to iron atom is from 4:1 to 1:4.

34. The composition of Claim 33 wherein the ratio of ligand to iron atom is from 2:1 to 1:2.

35. The composition of Claim 1 wherein the molar ratio of iron to cobalt is from 4:1 to 1:4.

36. The composition of Claim 35 wherein the molar ratio of iron to cobalt is from 2:1 to 1:3.

37. The composition of Claim 36 wherein the molar ratio of iron to cobalt is from 1:1 to 1:2.

38. The composition of claim 1 where the iron is ligated with a ligand selected from the group consisting of N-heterocycle, phosphine, and porphorine moieties.
39. The composition of Claim 1 wherein the cobalt is ligated with a ligand selected from the group consisting of N-heterocycle, phosphine, and porphorine moieties.
40. The composition of claim 1 wherein the cobalt compound is a cobalt carbonyl.
41. The composition of Claim 40 wherein the cobalt compound is dicobalt octacarbonyl.
42. The composition of Claim 41 wherein the the iron compound is iron pentacarbonyl.
43. The composition of Claim 40 wherein the cobalt compound is dicobalt octacarbonyl and the iron component is 2,3,7,8,12,13,17,18-octaethyl-21H, 23H-porphine iron(III) acetate.
44. The composition of Claim 43 wherein the molar ratio of Co:Fe:ligand is about 3:1:1.
45. The composition of Claim 40 wherein the cobalt carbonyl anion IR band is in the region 1875 to 1900 cm<sup>-1</sup>.
46. The composition of Claim 45 wherein the iron carbonyl IR band is in the region of 1950 to 2050 cm<sup>-1</sup>.
47. The composition of claim 40 wherein the iron carbonyl is selected from the group consisting of iron pentacarbonyl, diironnonacarbonyl, and triiron dodecacarbonyl.
48. The composition of claim 9 wherein the ligand is an aliphatic diimine.
49. A process for preparing 1,3-propanediol comprising the steps of:
- (a) contacting, in a reaction mixture, ethylene oxide, carbon monoxide, hydrogen, a non-reactive

reaction solvent, and a catalyst composition comprising:

(i) A cobalt compound; and

(ii) An iron component, optionally ligated with a ligand selected from the group consisting of N-heterocyclic, phosphine, and porphorine moieties, and;

(b) heating said mixture to a temperature of 30 to about 150°C and a pressure of at least 100 psi (690 kPa) for a time effective to produce a two-phase reaction product mixture comprising an upper phase comprising a major portion of the solvent, at least about 50 wt% of the catalyst composition, plus unreacted ethylene oxide, and a lower phase, which comprises a major portion of 1,3-propanediol.

50. The process of Claim 49 wherein the catalyst is made by the self-assembly method wherein all the components are brought together at the same time under synthesis gas conditions.

51. The process of Claim 49 wherein the catalyst is made stepwise wherein the iron compound is reacted with a ligand in the presence of syngas at a temperature in the range of 25 to 150°C and subsequently caused to undergo a redox reaction with the cobalt compound at a temperature in the range of 25 to 150°C.

52. The process of claim 49 wherein the hydrogen to carbon monoxide molar ratio is from 1:1 to 8:1.

53. The process of claim 52 wherein the hydrogen to carbon monoxide molar ratio is from 2:1 to 6:1.